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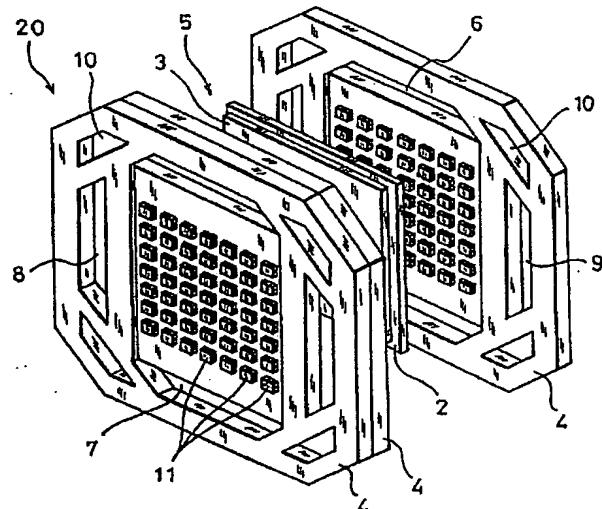
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(54) SEPARATEUR POUR PILE A COMBUSTIBLE ET METHODE DE FABRICATION DUDIT SEPARATEUR  
(54) SEPARATOR FOR A FUEL CELL AND A METHOD OF PRODUCING THE SAME

(57) In a separator for a fuel cell and a method of producing a separator for a fuel cell according to the invention, bond- carbon is used in which composition ratios are set to 60 to 90 wt.% (preferably, 70 to 87 wt.%) of graphite powder having an average diameter in a range of 15 to 125 .mu.m (preferably, 40 to 100 .mu.m) and 10 to 40 wt.% (preferably, 13 to 30 wt.%) of a thermosetting resin. The compound is previously cold-molded into a shape similar to a final molded shape. The preliminary molded member is then placed in a mold, and then molded into a separator of the final shape by applying a pressure of a range of 10 to 100 MPa. The surface roughness Ra of at least a portion of the separator contacting with an electrode is set to a range of 0.1 to 0.5 .mu.m. According to this configuration, fluidity and moldability are excellent, the contact resistance can be set to a value lower than a requested value while ensuring strength sufficient for preventing the separator from suffering a damage such as a break- age due to vibrations or the like, and the low contact resistance can be stably maintained.





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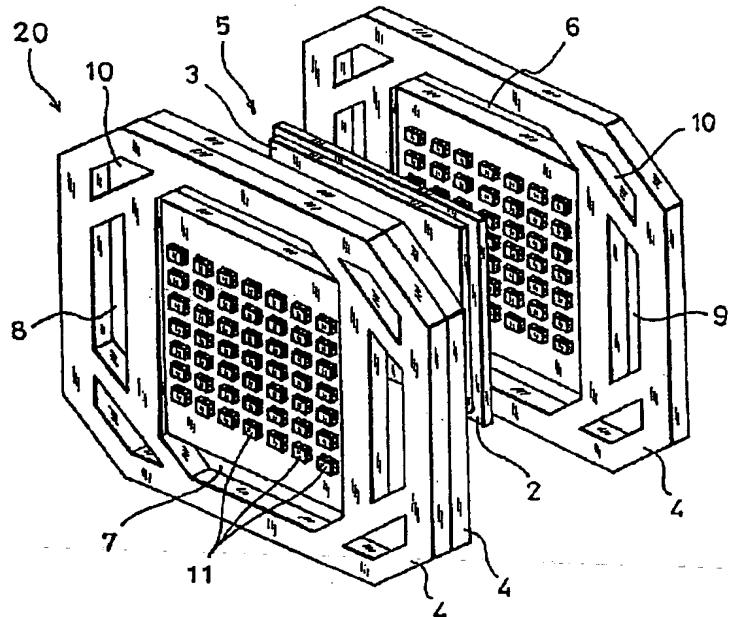
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(54) Titre : SEPARATEUR POUR PILE A COMBUSTIBLE ET METHODE DE FABRICATION DUDIT SEPARATEUR  
(54) Title: SEPARATOR FOR A FUEL CELL AND A METHOD OF PRODUCING THE SAME



(57) Abrégé/Abstract:

In a separator for a fuel cell and a method of producing a separator for a fuel cell according to the invention, bond-carbon is used in which composition ratios are set to 60 to 90 wt.% (preferably, 70 to 87 wt.%) of graphite powder having an average diameter in a range of 15 to 125  $\mu\text{m}$  (preferably, 40 to 100  $\mu\text{m}$ ), and 10 to 40 wt.% (preferably, 13 to 30 wt.%) of a thermosetting resin. The compound is previously cold-molded into a shape similar to a final molded shape. The preliminary molded member is then placed in a mold, and then molded into a separator of the final shape by applying a pressure of a range of 10 to 100 MPa. The surface roughness  $R_a$  of at least a portion of the separator contacting with an electrode is set to a range of 0.1 to 0.5  $\mu\text{m}$ . According to this configuration, fluidity and moldability are excellent, the contact resistance can be set to a value lower than a requested value while ensuring strength sufficient for preventing the separator from suffering a damage such as a breakage due to vibrations or the like, and the low contact resistance can be stably maintained.

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Abstract of the Disclosure

In a separator for a fuel cell and a method of producing a separator for a fuel cell according to the invention, bond-carbon is used in which composition ratios are set to 60 to 5 90 wt.% (preferably, 70 to 87 wt.%) of graphite powder having an average diameter in a range of 15 to 125  $\mu\text{m}$  (preferably, 40 to 100  $\mu\text{m}$ ), and 10 to 40 wt.% (preferably, 13 to 30 wt.%) of a thermosetting resin. The compound is previously cold-molded into a shape similar to a final molded shape. The 10 preliminary molded member is then placed in a mold, and then molded into a separator of the final shape by applying a pressure of a range of 10 to 100 MPa. The surface roughness Ra of at least a portion of the separator contacting with an electrode is set to a range of 0.1 to 0.5  $\mu\text{m}$ . According to 15 this configuration, fluidity and moldability are excellent, the contact resistance can be set to a value lower than a requested value while ensuring strength sufficient for preventing the separator from suffering a damage such as a breakage due to vibrations or the like, and the low contact resistance can be stably maintained.

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Title of the Invention

Separator for a fuel cell and a method of producing the  
same

5 Background of the Invention

1. Field of the Invention

The present invention relates to a separator for a fuel cell which is mainly used as a cell for an electric vehicle, and also to a method of producing the separator, and more 10 particularly to a separator for a fuel cell of the electrolyte type or the phosphoric acid type, and also to a method of producing the separator. In a fuel cell of such a type, a unit cell which is a unit constituting the cell is configured by: sandwiching a gas diffusion electrode having a sandwich 15 structure wherein an electrolyte membrane is configured by an ion exchange membrane, between an anode and a cathode; sandwiching the gas diffusion electrode between separators; and forming fuel gas passages and oxidant gas passages between the separators, and the anode and the cathode.

20

2. Description of the Prior Art

In a fuel cell, a fuel gas containing hydrogen is supplied to an anode, and an oxidant gas containing oxygen is supplied to a cathode, so that, in the anode and the cathode, 25 electrochemical reactions indicated by the formulae:

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occur, and, in the whole of the cell, an electrochemical reaction indicated by the formula:



proceeds. The chemical energy of the fuel is directly converted into an electrical energy, with the result that the cell can exert predetermined performance.

10 A separator for a fuel cell of the electrolyte type or the phosphoric acid type in which such energy conversion is conducted is requested to be gas-impermeable, and also to be made of an electrically conductive material. Conventionally, it is known that, as a material meeting the requirements, an electrically conductive resin is used. An electrically conductive resin is a complex which is configured by bonding graphite (carbon) powder by means of a thermosetting resin such as phenol resin, or a so-called bondcarbon (resin-bonded carbon) compound. A separator for a fuel cell is configured by forming such a bondcarbon compound into a predetermined 15 shape.

20

Conventionally, a separator for a fuel cell having a predetermined shape is formed by using such a bondcarbon compound in the following manner. With respect to the composition ratio of a thermosetting resin such as phenol resin and 25 graphite powder, 25 to 60 wt.% of the thermosetting resin is

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used as an adequate content in consideration of fluidity, moldability, and gas-impermeability of the bondcarbon compound, and in order to ensure the strength (compression and bending) sufficient for preventing the separator from suffering a damage such as a breakage due to vibrations or the like which may be produced during an handling operation in an assembling step of a unit cell of a fuel cell, or a use in an automobile.

In a conventional separator for a fuel cell which is configured by using a bondcarbon compound of such composition ratios, the content of a thermosetting resin serving as an electrically insulating material is large, and hence the conductivity of the separator itself is lowered so that the electrical resistance is increased. This is not preferable from the viewpoint of the performance of a fuel cell.

In order to improve the conductivity of a separator for a fuel cell which is configured by using a bondcarbon compound, it has been contemplated that the content of a thermosetting resin is reduced as far as possible. When the content of a thermosetting resin is reduced, however, elongation and fluidity of the bondcarbon compound during a molding process are lowered to impair the moldability, and the strength is low. When the resin content is 10 wt.% or less, particularly, the strength of a separator becomes insufficient, and therefore the separator easily suffers a damage such as a breakage

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or a crack due to vibrations or the like which are continuously applied to the separator in the case where the separator is used in an automobile.

By contrast, in the case where the resin content is set 5 to the above-mentioned adequate range (25 to 60 wt.%), elongation and fluidity of a bondcarbon compound are excellent and moldability is higher, and strength sufficient for preventing a separator from suffering a damage such as a breakage or a crack due to vibrations or the like can be ensured. However, 10 the contact resistance with respect to an electrode and serving as the primary factor which largely affects the performance of a fuel cell becomes higher, as the resin content is larger. When the resin content is larger than 40 wt.%, particularly, the contact resistance is suddenly increased, and 15 the performance of a fuel cell is extremely lowered.

The contact resistance serving as the primary factor which largely affects the performance of a fuel cell will be considered. Even when a fuel cell is used in an automobile in which vibrations are always applied to the fuel cell, the 20 contact resistance is requested to be stably maintained to 10  $\text{m}\Omega \cdot \text{cm}^2$  or lower. When the contact resistance is to be stably maintained to such a requested value, a countermeasure in which only the composition ratios of a thermosetting resin and graphite powder are considered cannot satisfy both the requirements on fluidity and moldability of a compound and the 25

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strength of a molded member (separator), and the contact resistance, as described above. Development of a separator for a fuel cell which is excellent in moldability and strength, and which can be stably maintained to a low contact resistance of 5  $10 \text{ m}\Omega \cdot \text{cm}^2$  or lower is strongly requested. At present, however, there exists no separator which can satisfy the request.

Summary of the Invention

10 The present invention has been conducted in order to satisfy the request. It is an object of the invention to provide a separator for a fuel cell which is excellent in fluidity and moldability, and in which, while ensuring strength sufficient for preventing the separator from suffering 15 a damage such as a breakage due to vibrations or the like, the contact resistance can be set to a value lower than a requested value, and the low contact resistance can be stably maintained.

It is another object of the invention to provide a method 20 of producing a separator for a fuel cell wherein, even when a molding material of low fluidity is used, a separator which has a uniform and correct shape, and in which a low contact resistance can be stably maintained can be surely produced.

In order to attain the object, the separator for a fuel 25 cell of the invention is a separator for a fuel cell consist-

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ing of a complex which is configured by bonding graphite powder by means of a thermosetting resin, and characterized in that, in the complex, a composition ratio of the graphite powder is set to 60 to 90 wt.%, a composition ratio of the thermosetting resin is set to 10 to 40 wt.%, and an average particle diameter of the graphite powder is set to a range of 5 15 to 125  $\mu\text{m}$ .

In the complex, preferably, the composition ratio of the graphite powder is set to 70 to 87 wt.%, and the composition 10 ratio of the thermosetting resin is set to 13 to 30 wt.%. Preferably, the average particle diameter of the graphite powder is set to a range of 40 to 100  $\mu\text{m}$ .

In order to meet the above-mentioned demands for development, the inventors of the invention have conducted intensive 15 studies on a separator for a fuel cell which is configured by using a bondcarbon compound, and finally found that the contact resistance serving as the primary factor which largely affects the performance of a fuel cell is determined not only by the composition ratios of a resin and graphite powder, the 20 average diameter of the graphite powder closely affects the performance at the highest degree, the contact resistance is largely varied depending on the size of the average diameter, and the average diameter of the graphite powder is closely related also to fluidity, moldability, and strength of the 25 compound. Based on this finding, the composition ratios of

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a resin and graphite powder, and the average diameter of the graphite powder have been respectively set to the above-mentioned ranges, thereby completing the invention.

According to the thus configured invention, as the graphite powder which is the one composition of the complex and which affects the contact resistance at the highest degree, graphite powder in which the average diameter is set to a range of 15 to 125  $\mu\text{m}$ , preferably, 40 to 100  $\mu\text{m}$  is used, the composition ratio of the thermosetting resin which is the other composition of the complex, and which largely affects the fluidity, the moldability, and the strength is set to a range of 10 to 40 wt.%, preferably, 13 to 30 wt.%, thereby attaining an effect that, while the complex serving as a molding material has excellent elongation and fluidity and exerts high moldability, and strength sufficient for preventing the separator from suffering a damage such as a breakage or a crack due to vibrations or the like can be ensured, the contact resistance with respect to an electrode can be set to a low value of  $10 \text{ m}\Omega\cdot\text{cm}^2$  or lower which is required in a separator for a fuel cell, and the low contact resistance can be stably maintained so that the performance of a fuel cell can be remarkably improved.

In the case where the average particle diameter of graphite powder is smaller than the above-mentioned range, or, for example, 10  $\mu\text{m}$  or smaller, the contact resistance is higher

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or  $15 \text{ m}\Omega \cdot \text{cm}^2$  or more, even when the resin content is adjusted to any value. Namely, the obtained contact resistance is very different from the value ( $10 \text{ m}\Omega \cdot \text{cm}^2$  or lower) which is required in a fuel cell to be used under conditions where vibrations are applied, such as the case of mounting on an automobile. In the case where the resin content is smaller than 10 wt.%, and also in the case where the average diameter of graphite powder is, for example,  $150 \mu\text{m}$  or more, i.e., exceeds the above-mentioned range, fluidity and moldability are improved, but a large number of breakages, minute cracks, and the like are produced by vibrations in edges of projections serving as contact faces with respect to an electrode. Even when a low contact resistance is obtained in an early stage of use, the contact resistance is suddenly increased after use of a short time, so that a low contact resistance meeting the above-mentioned demands cannot be maintained. This will be described later in detail.

In the separator for a fuel cell of the invention, when a surface roughness of a portion contacting with an electrode is set to a range of  $Ra = 0.1$  to  $0.5 \mu\text{m}$  as measured by a surface roughness meter having a probe of a diameter of  $5 \mu\text{m}$ , the contact resistance can be further lowered, so that further improvement of the performance of a cell can be attained.

The method of producing a separator for a fuel cell according to the invention is a method of producing a separator

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for a fuel cell configured by molding a complex in which composition ratios are set to 60 to 90 wt.% of graphite powder, and 10 to 40 wt.% of a thermosetting resin, and an average diameter of the graphite powder is set to a range of 15 to 125 <sup>5</sup>  $\mu\text{m}$ , and characterized in that the complex is previously cold-molded into a shape similar to a final molded shape by a pressure of a range of 2 to 10 MPa, the preliminary molded member is then placed in a mold, and the preliminary molded member is molded into the final shape by applying a pressure of a <sup>10</sup> range of 10 to 100 MPa.

Preferably, in the complex, the composition ratio of the graphite powder is set to 70 to 87 wt.%, the composition ratio of the thermosetting resin is set to 13 to 30 wt.%, and the average particle diameter of the graphite powder is set to a <sup>15</sup> range of 40 to 100  $\mu\text{m}$ .

The shape similar to a final molded shape means that the dimensions other than those in the direction of the molding pressure are similar to corresponding ones of the final molded member. Preferably, dimensions of the preliminary molded member in the direction of the molding pressure are set to be <sup>20</sup> about 1.0 to about 2.0 times dimensions of the final molded member. When such a preliminary molded member is used, the mold density and the volume resistivity can be further improved.

<sup>25</sup> According to the production method of the invention hav-

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ing the above-described molding means, the two-step molding is employed wherein a complex (bondcarbon compound) is previously cold-molded into a shape similar to the final molded shape by a pressure of a range of 2 to 10 MPa, and the preliminary molded member is placed in a mold and then molded into the final shape by applying a high molding pressure of a range of 10 to 100 MPa. Even when a complex (molding material) which is low in elongation and fluidity is used, therefore, the compound can surely extend to every corner of the mold so that, while suppressing molding unevenness, the mold density is increased and the complex can be charged more uniformly. As a result, it is possible to surely and easily obtain a uniform separator which exhibits a low contact resistance and has a good conductivity, and which is uniform, and is correct also in shape.

As the thermosetting resin which is useful in the invention, phenol resin which is excellent in wettability with respect to graphite powder may be most preferably used. Alternatively, any other resin such as polycarbodiimide resin, 20 epoxy resin, furfuryl alcohol resin, urea resin, melamine resin, unsaturated polyester resin, or alkyd resin may be used as far as the resin causes a thermosetting reaction when the resin is heated, and is stable against the operating temperature of the fuel cell and components of the supplied gasses.

25 As the graphite powder which is useful in the invention,

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powder of graphite of any kind, including natural graphite, artificial graphite, carbon black, kish graphite, and expanded graphite may be used. In consideration of conditions such as the cost, the kind can be arbitrarily selected. In the case 5 where expanded graphite is used, particularly, a layer structure is formed by expanding the volume of the graphite as a result of heating. When the molding pressure is applied, layers can twine together to be firmly bonded to one another. Therefore, expanded graphite is effective in a complex in 10 which the ratio of a thermosetting resin is to be reduced.

Other objects and effects of the invention will be clarified in embodiments which will be described below.

#### Brief Description of the Drawings

15 Fig. 1 is an exploded perspective view showing the configuration of a stack structure constituting a solid polymer electrolyte type fuel cell which has the separator of the invention;

Fig. 2 is an external front view of the separator in the 20 solid polymer electrolyte type fuel cell;

Fig. 3 is an enlarged section view of main portions and showing the configuration of a unit cell which is a unit constituting the solid polymer electrolyte type fuel cell;

Fig. 4A is a view illustrating a step of producing the 25 separator, and Fig. 4B is a view illustrating the manner of

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the production;

Fig. 5 is a perspective view illustrating specifications of a test piece;

Fig. 6 is an enlarged section view of portion A which is 5 circled in Fig. 5; and

Fig. 7 is a graph showing correlation between the resin content and the compressive strength of embodiments and comparison examples.

10 Preferred Embodiments of the Invention

Hereinafter, embodiments of the invention will be described with reference to the accompanying drawings.

First, the configuration and the operation of a solid polymer electrolyte type fuel cell having the separator of the invention will be briefly described with reference to Figs. 15 1 to 3.

The solid polymer electrolyte type fuel cell 20 has a stack structure in which plural unit cells 5 are stacked and collector plates (not shown) are respectively placed on both 20 the ends. Each of the unit cells 5 is configured by: an electrolyte membrane 1 which is an ion exchange membrane made of, for example, a fluororesin; an anode 2 and a cathode 3 which are formed by carbon cloth woven of carbon filaments, carbon paper, or carbon felt, and which sandwich the electrolyte 25 membrane 1 to constitute a gas diffusion electrode having a

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sandwich structure; and separators 4 which sandwich the sandwich structure.

In each of the separators 4, as shown in Fig. 2, fuel gas holes 6 and 7 for a fuel gas containing hydrogen, oxidant gas holes 8 and 9 for an oxidant gas containing oxygen, and a coolant water hole 10 are formed in the peripheral area. When plural unit cells 5 are stacked, the holes 6, 7, 8, 9, and 10 of the separators 4 of the unit cells constitute holes passing through the fuel cell 20 in the longitudinal direction to form a fuel gas supply manifold, a fuel gas discharge manifold, an oxidant gas supply manifold, an oxidant gas discharge manifold, and a coolant water passage, respectively.

As shown in Fig. 3, a large number of ribs 11 having a predetermined shape are protrudingly formed on the surfaces of the separators 4 which sandwich the electrolyte membrane 1, the anode 2, and the cathode 3. Fuel gas passages 12 are formed between the ribs 11 of one of the separators 4 and the surface of the anode 2. Oxidant gas passages 13 are formed between the ribs 11 of the other separator 4 and the surface 20 of the cathode 3.

In the solid polymer electrolyte type fuel cell 20 configured as a stack structure in which plural unit cells 5 are stacked and the collector plates are respectively placed on both the ends, the fuel gas which is supplied from an external fuel gas supplying device to the fuel cell 20, and which con-

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tains hydrogen is then supplied into the fuel gas passages 12 of each unit cell 5 via the fuel gas supply manifold to cause the electrochemical reaction indicated by formula (1) above, on the side of the anode 2 of the unit cell 5. After the 5 reaction, the fuel gas is discharged to the outside via the fuel gas passages 12 of the unit cell 5 and the fuel gas discharge manifold. At the same time, the oxidant gas (air) which is supplied from an external oxidant gas supplying device to the fuel cell 20, and which contains oxygen is then 10 supplied into the oxidant gas passages 13 of each unit cell 5 via the oxidant gas supply manifold to cause the electrochemical reaction indicated by formula (2) above, on the side of the cathode 3 of the unit cell 5. After the reaction, the oxidant gas is discharged to the outside via the oxidant gas 15 passages 13 of the unit cell 5 and the oxidant gas discharge manifold.

In accordance with the electrochemical reactions of formulae (1) and (2) above, in the whole of the fuel cell 20, the electrochemical reaction indicated by the formula (3) proceeds, so that the chemical energy of the fuel is directly 20 converted into an electrical energy, with the result that the cell can exert predetermined performance. Because of the characteristics of the electrolyte membrane 1, the fuel cell 25 is operated in a temperature range of about 80 to 100°C, and hence involves heat generation. During operation of the

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fuel cell 20, therefore, coolant water is supplied from an external coolant water supplying device to the fuel cell 20, and the coolant water is circulated through the coolant water passage, thereby preventing the temperature of the interior 5 of the fuel cell 20 from being raised.

Each of the separators 4 in the solid polymer electrolyte type fuel cell 20 which is configured and operates as described above is produced in the following manner. A method of producing the separator will be described with reference 10 to Figs. 4A and 4B. The separator 4 is molded by using a complex (bondcarbon) in which the composition ratios are set to 60 to 90 wt.%, preferably, 70 to 87 wt.% of graphite powder, and 10 to 40 wt.%, preferably, 13 to 30 wt.% of a thermosetting resin. The graphite powder and the thermosetting 15 resin are uniformly mixed with each other and adjusted to produce a predetermined compound (step S100). While applying a pressure in a range of 2 to 10 MPa to the compound, the compound is previously cold-molded into a shape similar to a final molded shape (step S101). As shown in Fig. 4B, the 20 preliminary molded member is then placed in a mold 14 having a predetermined final shape (step S102). Under this state, the mold 14 is heated to 150 to 170°C, and a pressing machine which is not shown is operated to apply a pressure in a range 25 of 10 to 100 MPa, preferably, 20 to 50 MPa in the direction of the arrow f in Fig. 4B (step S103), thereby producing the

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separator 4 having the final shape which corresponds to the shape of the mold 14 (step S104).

In the separator 4 which is produced as described above, with respect to the composition ratios of the bondcarbon constituting the separator 4, the amount of the thermosetting resin is as small as 10 to 40 wt.% (preferably, 13 to 30 wt.%), and hence the bondcarbon itself has a high conductivity. After the compound of the bondcarbon is preliminary molded into a shape similar to the final molded shape, the preliminary molded member is placed in the mold 14, and a high molding pressure of 10 to 100 MPa (preferably, 20 to 50 MPa) is then applied to the member while heating the mold to 150 to 170°C. Therefore, the thermosetting resin melts and a thermosetting reaction occurs, with the result that the preliminary molded member can be uniformly molded into the separator 4 in which the mold density is high, and which has a predetermined shape.

As the graphite powder which affects the contact resistance at the highest degree, graphite powder in which the average diameter is set to a range of 15 to 125  $\mu\text{m}$ , preferably, 40 to 100  $\mu\text{m}$  is used, and the composition ratio of the thermosetting resin which largely affects the fluidity, the moldability, and the strength is set to a range of 10 to 40 wt.%, preferably, 13 to 30 wt.%. As a result, while the complex serving as a molding material has excellent elongation

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and fluidity and exerts high moldability, and strength sufficient for preventing the separator from suffering a damage such as a breakage due to vibrations or the like can be ensured, the contact resistance with respect to an electrode can 5 be set to a low value of  $10 \text{ m}\Omega \cdot \text{cm}^2$  or lower.

Hereinafter, the invention will be described in more detail by way of embodiments.

**<Embodiments 1 to 4>**

Bondcarbon compounds of powders of natural graphite 10 (products of SEC Co. Ltd.) respectively having average particle diameters of 15  $\mu\text{m}$ , 45  $\mu\text{m}$ , 100  $\mu\text{m}$ , and 125  $\mu\text{m}$ , and phenol resin were prepared at the composition ratios listed in Table 1. Each of the compounds was charged into a mold. A molding pressure of 15 MPa was applied to the compound for 2 minutes 15 at a molding temperature of 160°C. Thereafter, the compound was heated to 170°C for 30 minutes, thereby molding a test piece TP in which, as shown in Fig. 5, width (a)  $\times$  length (b)  $\times$  thickness (t) is 170  $\times$  230  $\times$  2 (mm), and, as shown in Fig. 6, gas passages R where depth (d)  $\times$  width (w) is 1  $\times$  2 (mm) 20 are formed in parallel. In each of the test pieces TP of Embodiments 1 to 4, the surface roughness (Ra) was measured at arbitrary 10 points by a surface roughness meter having a probe of a diameter of 5  $\mu\text{m}$ , and in accordance with the method specified in JIS B 0601-1994. The results are in the ranges 25 listed in Table 1.

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<Comparison Examples 1 to 6>

Bondcarbon compounds of powders of natural graphite (products of SEC Co. Ltd.) respectively having average particle diameters of 10  $\mu\text{m}$ , 15  $\mu\text{m}$ , 45  $\mu\text{m}$ , 100  $\mu\text{m}$ , 125  $\mu\text{m}$ , and 150  $\mu\text{m}$ , and phenol resin were prepared at the composition ratios listed in Table 1. The compounds were molded in the same molding conditions as Embodiments 1 to 4, into test pieces TP of the shapes shown in Figs. 5 and 6. In each of the test pieces TP of Comparison examples 1 to 6, the surface roughness (Ra) was measured in the same manner as described above. The results are in the ranges listed in Table 1.

The contact resistance of each of the test pieces TP of Embodiments 1 to 4 and Comparison examples 1 to 6 was measured. The results are listed in Table 1. In each of the test pieces TP of Embodiments 1, 3, and 4 and Comparison examples 1 and 6, the compressive strength was measured, and the results shown in Fig. 7 were obtained. In each pair of test pieces TP which are equal to each other in average diameter of graphite powder, namely, Embodiment 1 and Comparison example 2 (15  $\mu\text{m}$ ), Embodiment 3 and Comparison example 4 (100  $\mu\text{m}$ ), and Embodiment 4 and Comparison example 5 (125  $\mu\text{m}$ ), the compressive strengths are substantially equal to each other.

In each of Embodiments 1 to 4 and Comparison examples 1 and 6, ten test pieces TP were molded. A vibration test was conducted so that vibrations of 1,200 cycles/minute and an

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amplitude of 16  $\mu\text{m}$  were applied to the test pieces TP. After the vibration test, the appearance of each test piece TP was observed, and the number of non-defective test pieces in which breakage or crack is not produced in, for example, edges of 5 projections for forming the gas passages was counted. The results are listed in Table 2.

Table 1

|                         | RESIN<br>CONTENT<br>(VOL. %)→                           | 1 0  | 1 5 | 3 0 | 4 0 | 5 0  |   |
|-------------------------|---|--|-----|-----|-----|------|---|
|                         | AVERAGE PARTICLE<br>DIAMETER OF<br>GRAPHITE POWDER<br>↓ | ↓ CONTACT RESISTANCE( $\text{m}\Omega \cdot \text{cm}^2$ ) |     |     |     |      | SURFACE<br>ROUGHNESS $\text{Ra}$<br>↓ ( $\mu\text{m}$ ) |
| EMBODIMENT 1            | 15 $\mu\text{m}$  | 7.9  | 8.4 | 8.8 | 9.1 | 14.0 | 0.1~0.5   |
| EMBODIMENT 2            | 45 $\mu\text{m}$  | 4.7  | 5.1 | 5.6 | 6.4 | 12.6 | 0.1~0.5   |
| EMBODIMENT 3            | 100 $\mu\text{m}$                                       | 3.1  | 3.2 | 3.8 | 5.6 | 12.1 | 0.1~0.5   |
| EMBODIMENT 4            | 125 $\mu\text{m}$                                       | 2.8  | 3.0 | 4.6 | 5.3 | 11.9 | 0.1~0.5   |
| COMPARISON<br>EXAMPLE 1 | 10 $\mu\text{m}$  | 16   | 32  | 39  | 43  | 60   | 1.2~1.9   |
| COMPARISON<br>EXAMPLE 2 | 15 $\mu\text{m}$  | 8.7  | 9.2 | 9.6 | 9.9 | 17.0 | 0.9~1.8   |
| COMPARISON<br>EXAMPLE 3 | 45 $\mu\text{m}$  | 6.9  | 7.2 | 7.4 | 7.8 | 14.8 | 0.8~2.1   |
| COMPARISON<br>EXAMPLE 4 | 100 $\mu\text{m}$                                       | 4.6  | 5.6 | 6.7 | 8.9 | 13.2 | 0.8~1.7   |
| COMPARISON<br>EXAMPLE 5 | 125 $\mu\text{m}$                                       | 3.1  | 3.4 | 5.8 | 7.5 | 14.2 | 1.2~1.8   |
| COMPARISON<br>EXAMPLE 6 | 150 $\mu\text{m}$                                       | 2.6  | 2.8 | 3.9 | 5.0 | 13.2 | 1.2~1.8   |

Table 2

|                         | RESIN<br>CONTENT<br>(VOL. %)→                           | 1 0   | 1 5 | 3 0 | 4 0 | 5 0 |
|-------------------------|---|---|-----|-----|-----|-----|
|                         | AVERAGE PARTICLE<br>DIAMETER OF<br>GRAPHITE POWDER<br>↓ | ↓ NUMBER OF NON-DEFECTIVE ONES OF 10<br>TEST PIECES |     |     |     |     |
| EMBODIMENT 1            | 15 $\mu\text{m}$  | 7   | 1 0 | 1 0 | 1 0 | 1 0 |
| EMBODIMENT 2            | 45 $\mu\text{m}$  | 6   | 1 0 | 1 0 | 1 0 | 1 0 |
| EMBODIMENT 3            | 100 $\mu\text{m}$                                       | 7   | 1 0 | 1 0 | 1 0 | 1 0 |
| EMBODIMENT 4            | 125 $\mu\text{m}$                                       | 7   | 1 0 | 1 0 | 1 0 | 1 0 |
| COMPARISON<br>EXAMPLE 1 | 10 $\mu\text{m}$  | 6   | 8   | 8   | 1 0 | 1 0 |
| COMPARISON<br>EXAMPLE 6 | 150 $\mu\text{m}$                                       | 7   | 7   | 8   | 8   | 7   |

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As apparent from the results listed in Table 1, in Comparison example 1 in which the average particle diameter of graphite powder is smaller than 10  $\mu\text{m}$ , the contact resistance is not lower than  $15 \text{ m}\Omega\cdot\text{cm}^2$  even when the resin content is 5 adjusted to any value, or namely is very different from the value ( $10 \text{ m}\Omega\cdot\text{cm}^2$  or lower) which is required in a separator for a fuel cell. By contrast, in Embodiments 1 to 4 and Comparison examples 2 to 5 in which the average particle diameter of graphite powder is 15 to 125  $\mu\text{m}$ , when the resin content is 10 set to a range of 10 to 40 wt.%, the contact resistance can be set so as not to be higher than  $10 \text{ m}\Omega\cdot\text{cm}^2$ , but, when the resin content is set to 50 wt.%, the contact resistance is 11  $\text{m}\Omega\cdot\text{cm}^2$  or higher, or cannot be set to be lower than the required value. Furthermore, it was confirmed that, even in the 15 case where the average diameter of graphite powder is in a range of 15 to 125  $\mu\text{m}$  and the resin content is in a range of 10 to 40 wt.%, in Comparison examples 2 to 5 in which the surface roughness  $\text{Ra}$  is 0.6  $\mu\text{m}$  or more, the contact resistance is higher by 0.8 to  $2.23 \text{ m}\Omega\cdot\text{cm}^2$  than Embodiments 1 to 4 in 20 which the surface roughness  $\text{Ra}$  is in a range of 0.1 to 0.5  $\mu\text{m}$ .

As seen from the results of Fig. 7 and Table 2, it was confirmed that Comparison example 1 in which the resin content is smaller than 10 wt.%, and Comparison example 6 in which the average diameter of graphite powder is 150  $\mu\text{m}$  are defective 25 test pieces wherein minute breakages or cracks are produced

in edges of projections for forming the gas passages.

From the results of the tests, it was finally noted that the conditions for: attaining a low contact resistance (10  $\text{m}\Omega \cdot \text{cm}^2$  or lower) which is required in a separator for a fuel cell; and, even in a use under conditions where vibrations are applied, such as the case of mounting on an automobile, preventing breakages, cracks, or the like from occurring, and maintaining an initial low contact resistance are that the resin content is in a range of 10 to 40 wt.%, preferably, 13 to 30 wt.% and the average diameter of graphite powder is in a range of 15 to 125  $\mu\text{m}$ , preferably, 40 to 100  $\mu\text{m}$ . When the average diameter of graphite powder is set to a range of 40 to 100  $\mu\text{m}$  and the surface roughness  $\text{Ra}$  of a portion contacting with an electrode is set to a range of 0.1 to 0.5  $\mu\text{m}$ , the contact resistance can be further lowered, so that more improvement of the performance of a cell can be attained.

The entire disclosure of Japanese Patent Application No. 2000-183236 filed on June 19, 2000 including specification, claims, drawings and summary are incorporated herein by reference in its entirety.

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What is claimed is:

1. A separator for a fuel cell consisting of a complex which is configured by bonding graphite powder by means of a thermosetting resin, wherein

5 in said complex, a composition ratio of said graphite powder is set to 60 to 90 wt.%, a composition ratio of said thermosetting resin is set to 10 to 40 wt.%, and

an average particle diameter of said graphite powder is set to a range of 15 to 125  $\mu\text{m}$ .

10 2. A separator for a fuel cell according to claim 1, wherein, in said complex, the composition ratio of said graphite powder is set to 70 to 87 wt.%, and the composition ratio of said thermosetting resin is set to 13 to 30 wt.%.

15 3. A separator for a fuel cell according to claim 1, wherein the average particle diameter of said graphite powder is set to a range of 40 to 100  $\mu\text{m}$ .

4. A separator for a fuel cell according to claim 2, wherein the average particle diameter of said graphite powder is set to a range of 40 to 100  $\mu\text{m}$ .

20 5. A separator for a fuel cell according to claim 1, wherein a surface roughness of at least a portion contacting with an electrode is set to a range of  $\text{Ra} = 0.1$  to  $0.5 \mu\text{m}$  as measured by a surface roughness meter having a probe of a diameter of  $5 \mu\text{m}$ .

25 6. A separator for a fuel cell according to claim 2,

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wherein a surface roughness of at least a portion contacting with an electrode is set to a range of  $R_a = 0.1$  to  $0.5 \mu\text{m}$  as measured by a surface roughness meter having a probe of a diameter of  $5 \mu\text{m}$ .

5 7. A method of producing a separator for a fuel cell configured by molding a complex in which composition ratios are set to 60 to 90 wt.% of graphite powder, and 10 to 40 wt.% of a thermosetting resin, and an average particle diameter of said graphite powder is set to a range of 15 to 125  $\mu\text{m}$ ,

10 wherein

15 said complex is previously cold-molded by a pressure of a range of 2 to 10 MPa into a shape similar to a final molded shape, and

20 said preliminary molded member is then placed in a mold, and molded into the final shape by applying a pressure of 10 to 100 MPa.

8. A method of producing a separator for a fuel cell according to claim 7, wherein, in said complex, the composition ratio of said graphite powder is set to 70 to 87 wt.%, the composition ratio of said thermosetting resin is set to 13 to 30 wt.%, and the average particle diameter of said graphite powder is set to a range of 40 to 100  $\mu\text{m}$ .

25 9. A method of producing a separator for a fuel cell according to claim 7, wherein a surface roughness of at least a portion of said final molding member contacting with an

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electrode is set to a range of  $R_a = 0.1$  to  $0.5 \mu\text{m}$  as measured by a surface roughness meter having a probe of a diameter of  $5 \mu\text{m}$ .

10. A method of producing a separator for a fuel cell according to claim 8, wherein a surface roughness of at least a portion of said final molding member contacting with an electrode is set to a range of  $R_a = 0.1$  to  $0.5 \mu\text{m}$  as measured by a surface roughness meter having a probe of a diameter of  $5 \mu\text{m}$ .

Fig. 1

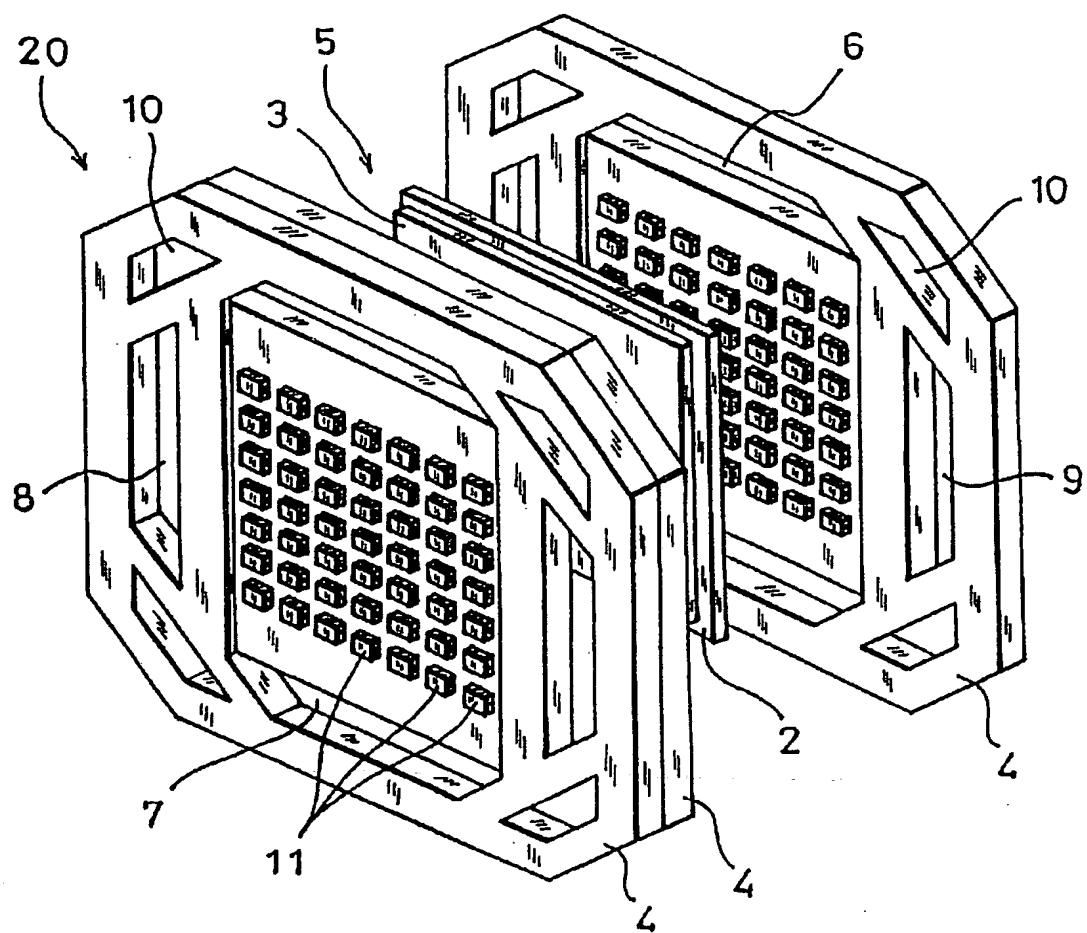


Fig. 2

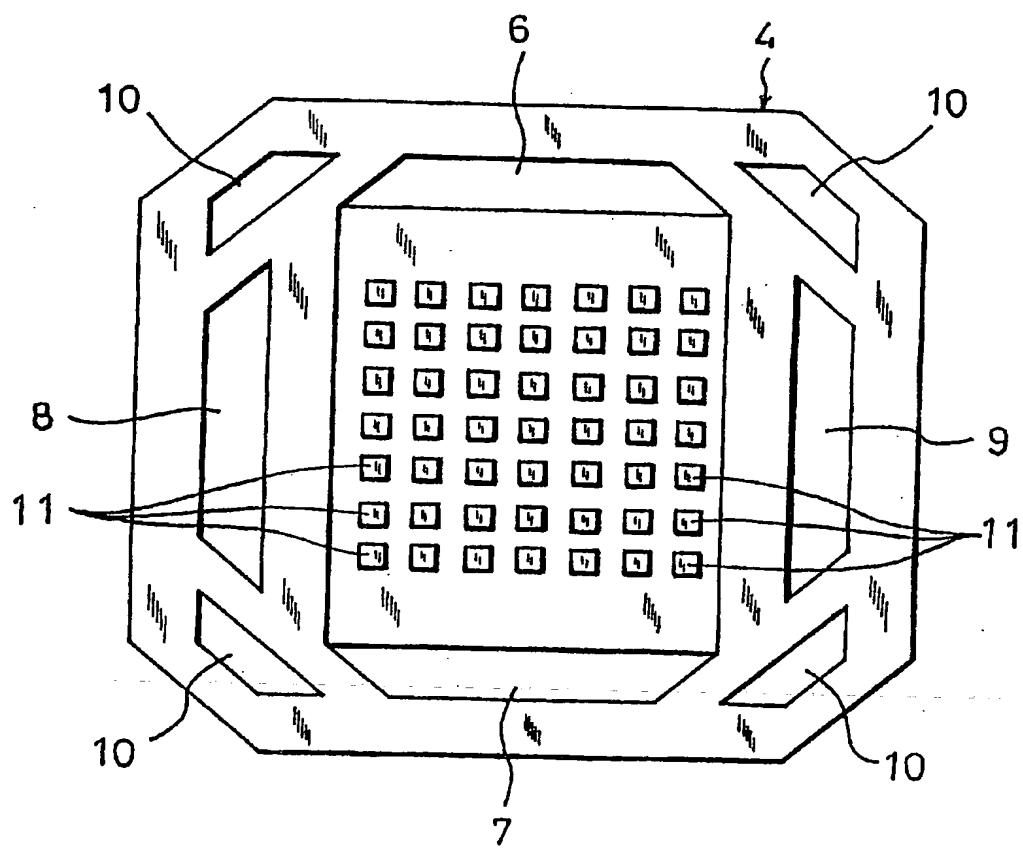


Fig. 3

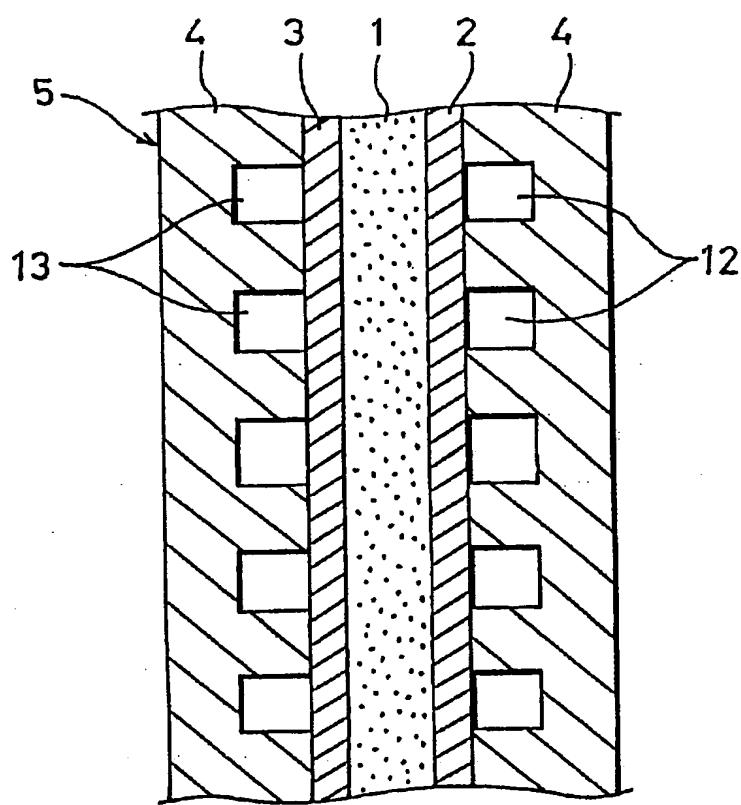


Fig. 4A

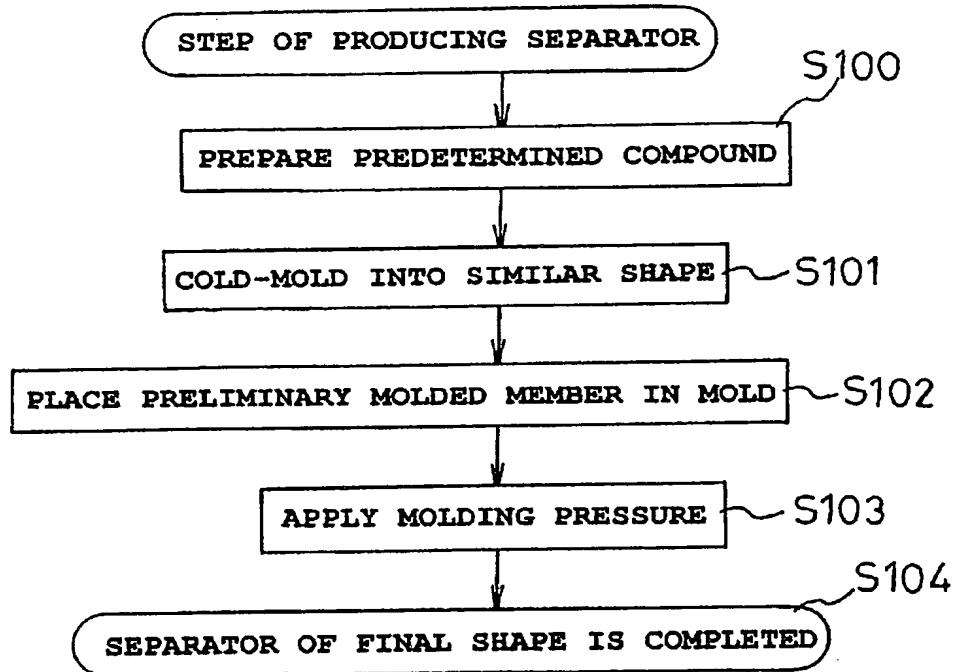


Fig. 4B

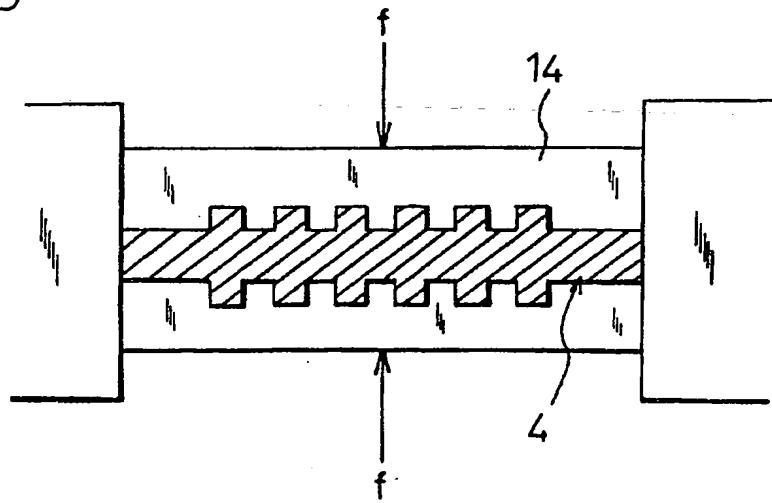


Fig. 5

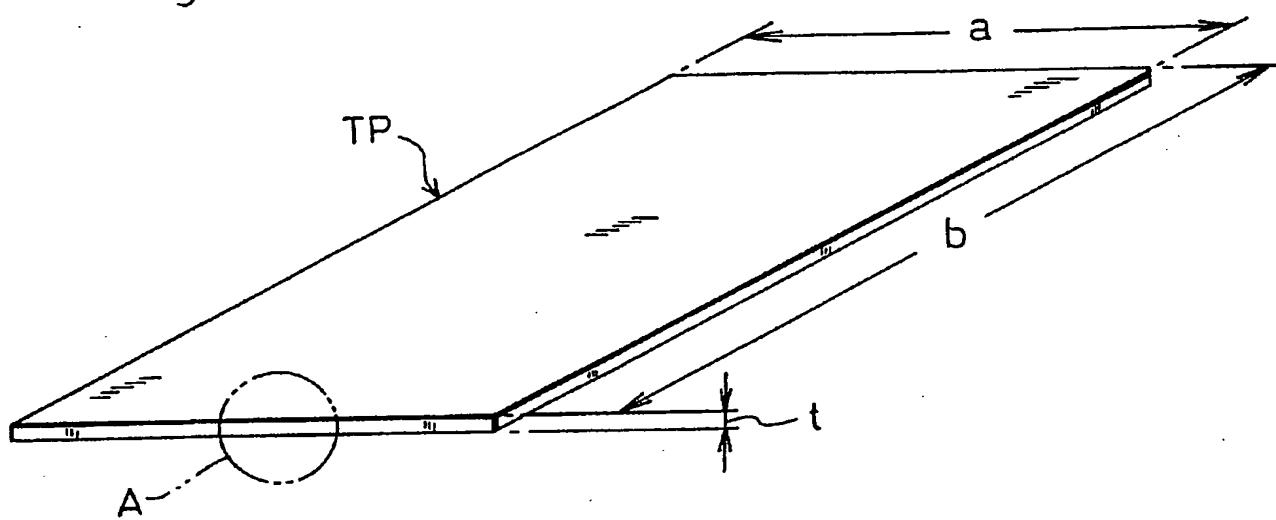


Fig. 6

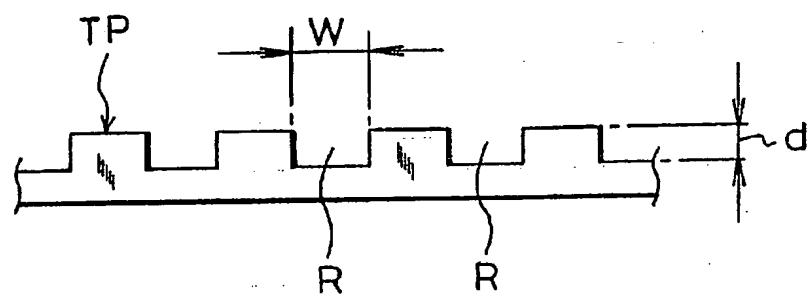


Fig. 7

